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Coexistence of resistive switching and negative differential resistance in -Fe2O3 nanorod film

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Date: 2017-08-18T00:00:00+00:00

Abstract

We report the coexistence of resistance switching (RS) behavior and negative differential resistance (NDR) phenomenon in -Fe2O3 nanorod film grown in situ on fluorine-doped tin oxide glass substrate. The reversible switching of the low- and high-resistance states (LRS and HRS, respectively) of the film device can be excited simply by applying bias voltage. The switching from HRS to LRS was initiated at the negative bias region, whereas the NDR process followed by the reversion of HRS occurred at the positive bias region. With the increase in compliant current (CC), the carrier conduction models of LRS and HRS both changed and the current-voltage (I-V) relationships at the NDR region were seriously affected by the thermal process according to the level of applied CC. The co-existence of RS and NDR was possibly caused by defects during migration, such as oxygen vacancies and interstitial iron ions, which were formed in the -Fe2O3 nanorod film. This work provided information on the ongoing effort toward developing novel electrical features of advanced transition metal oxide devices.

Full Text

Preamble

Coexistence of Resistance Switching and Negative Differential Resistance in -Fe O Nanorod

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Abstract

We report the coexistence of resistance switching (RS) behavior and negative differential resistance (NDR) phenomenon in an -Fe O nanorod film grown in situ on a fluorine-doped tin oxide glass substrate. The device can be reversibly switched between low-resistance (LRS) and high-resistance (HRS) states simply by applying a bias voltage. Switching from HRS to LRS occurs in the negative bias region, while the NDR process followed by a return to HRS takes place in the positive bias region. As the compliance current (CC) increases, the carrier conduction mechanisms for both LRS and HRS change, and the current–voltage (I–V) relationships in the NDR region become strongly influenced by thermal effects that depend on the applied CC level. The coexistence of RS and NDR is likely caused by migration-induced defects such as oxygen vacancies and interstitial iron ions within the -Fe O nanorod film. This work provides insights for ongoing efforts to develop novel electrical functionalities in advanced transition metal oxide devices.

Keywords: resistance switching, negative differential resistance, -Fe O nanorod film, defects, thermal process

Introduction

The application of transition metal oxides (TMOs) with resistance switching (RS) potential in solid-state memory technology has attracted considerable interest. While two basic models—bipolar and unipolar—describe the RS characteristics of these memories, unusual transport behaviors can still emerge in TMOs. Examples include the co-occurrence of bipolar and unipolar RS in ZnO[1] and the coincidence of memory and threshold RS in NiO.[2] These cases suggest that the RS features of TMOs have not been exhaustively defined. Another remarkable phenomenon is the coexistence of RS and negative differential resistance (NDR), which indicates an abrupt change in resistivity or conductivity. NDR typically occurs in III–V compound semiconductors or II–VI chalcogenide semiconductors according to intervalley carrier transfer theory.[3], It has been widely applied in various electronic devices such as resonant tunneling transistors, frequency multipliers,[3] and high-frequency oscillators. Consequently, developing this phenomenon to enhance the memory application potential of TMOs is highly anticipated, as reported in recent literature.[3] ,[31] However, the occurrence of NDR in TMOs and its correlation with RS behavior remain puzzling.

In TMOs, NDR often appears as an "N" -shaped feature in RS test results. The emergence of NDR indicates that current first increases with rising voltage before reaching a peak voltage (V), then drops to its lowest value at the valley point voltage (V), and finally increases again with continuously rising voltage. This phenomenon resembles the "Reset" step in RS (rapid switching from LRS to HRS). However, the current in NDR does not drop directly to HRS; instead, it decreases slowly before rising again, which can be viewed as an incomplete "Reset" process or an intermediate state between LRS and HRS.

Studies have shown that current can eventually revert to HRS following such an incomplete "Reset" process, enabling reproducible RS behavior across successive voltage sweeping cycles. Nevertheless, due to the substantial variability in TMO electrical properties induced by external means, the underlying mechanisms may differ significantly across materials.[32] Whether NDR truly benefits the stability and sustainability of RS behavior remains inconclusive.

An emerging issue of interest is that some crystallized TMOs, well known as nonvolatile memory candidates in their amorphous state, show potential for NDR to enable and enrich RS functions in nanodevice design. For instance, Ito et al. fabricated ZnO nanocrystal films with a double Schottky barrier structure to generate NDR that could tune the RS properties of single-electron devices.[1] Chuang et al. found that NDR in wurtzite ZnO nanorods can cause breakdown of high current with an on–off ratio up to 10 .[11] Additionally, Ziegler et al. demonstrated that crystalline TiO films can exhibit stable bipolar RS characteristics even at the atomic scale when coordinated with NDR.[12] Revealing the mechanism of NDR in various materials to clarify its correlation with RS behavior in TMOs has become extremely important. Several defect-related theories have been proposed, including resonant tunneling conduction of charges[13] and current from oxygen vacancy migration. These results show that NDR emergence in TMOs and its mechanism of action are closely related to the microscopic structures of the TMOs themselves.

-Fe O (hematite) is renowned for its low cost, high safety, easy preparation, and good stability, and it has been used in various electronic devices such as gas sensors[1] and field emitters.[1] Moreover, -Fe O is compatible with various substrates (Si, conductive glass, etc.),[1] making it a promising candidate material for advanced devices. While amorphous iron oxides (Fe O , FeO) typically exhibit unipolar RS characteristics,[1] ,[1] the current–voltage (I–V) properties of crystallized -Fe O have been limited to Mott insulator behavior,[1] and its electrical conductivity is unstable under changing external pressure or temperature. The influence of varied external biases on the internal crystal lattice structure has rarely been reported. A recent study demonstrated that Ga-doped -Fe O displays magnetic field-induced NDR,[2] but the characteristics of pure -Fe O film under varying external bias remain unknown.

This work presents the first report on the coexistence of RS and NDR behaviors in well-crystallized -Fe O nanorod film prepared through hydrothermal treatment of a fluorine-doped tin oxide (FTO) substrate in FeCl solution followed by annealing at 550 °C in air.[21] We investigated the impact of compliance current (CC) levels and voltage ranges on the I–V properties of the nanorod film. The synthesized film exhibited RS ON behavior in the negative bias region and NDR phenomenon in the positive bias region. In the NDR region, the current dropped drastically to nearly HRS under high CC levels, whereas it gently decreased to an intermediate state between HRS and LRS under low CC levels. We discuss the underlying mechanisms of NDR, which are closely related to inevitable defects in the grown film. This work represents pioneering research

on the special I–V properties associated with NDR in -Fe O nanorod film.

Experimental

The FTO glass substrate was placed in 25 mL of aqueous solution containing 0.05 mol/L FeCl and 0.25 mol/L NaNO for hydrothermal treatment at 120 °C for 3 h. The as-formed thin yellow-colored -FeOOH nanorod film on the substrate was washed with deionized water and ethyl alcohol using an ultrasonic washer to remove residual ions. The substrate was then annealed at 550 °C for 2 h to obtain the final -Fe O nanorod film. Field-emission scanning electron microscopy (FESEM, Sirion 200 FRG), transmission electron microscopy (TEM, JEOL JEM-2010), and X-ray diffraction (XRD, Philips X' pert Pro diffractometer with Cu K radiation) were used to observe the morphologies, distinguish fine structures, and determine crystalline information. Top tungsten (W) electrodes with 25 m diameter were deposited via small ion sputtering (SBC-12, KYKY, Beijing). Current–voltage (I–V) measurements were performed with a semiconductor analyzer (Agilent B1500A). Temperature-dependent resistance of the film was investigated using a four-probe method in a physical property measurement system (PPMS).

Results and Discussion

[Figure 1: see original paper] shows the basic characterizations of the as-formed -Fe O film. The top morphology in Fig. 1(a) reveals a compact nanorod film with an average diameter of approximately 50 nm; the inset shows an image of the actual specimen. Fig. 1(b) shows that the average film thickness on the FTO conductive glass substrate was approximately 420 nm. Fig. 1(e) displays a random nanorod, while Fig. 1(c) shows a high-resolution TEM image of the region within the white wire frame in Fig. 1(e). The corresponding selected area electron diffraction (SAED) pattern (Fig. 1(d)) provides consistent structural information. Three types of lattice fringes were observed with an interplanar spacing of 0.253 nm and angles of 60° between any two planes. These results match the relationships among the (110), (−120), and (2−10) planes, which belong to the {110} facets of the rhombohedral -Fe O structure. The XRD pattern (Fig. 1(f)) also confirms that the diffraction peaks are well indexed to rhombohedral hematite (JCPDS 33-0664), with the (110) plane as the second strongest diffraction peak, while the remaining peaks correspond to tin dioxide (SnO) (JCPDS 46-1088).

Fig. 1 Basic characterizations of the -Fe O nanorod film on FTO substrate: FESEM images of (a) top and (b) side morphologies. (c) High-resolution image corresponding to the white wire frame region in (e). (d) SAED pattern corresponding to (e). (e) TEM image of a single nanorod. (f) XRD pattern of the film.

[Figure 2: see original paper] shows five consecutive I–V sweeping curves under CC levels of 10 and 14 mA. The top W electrode received a positive bias at

the beginning of each sweep, following the sequence: V $\rightarrow 0 \rightarrow -$V $\rightarrow 0 \rightarrow$ V . At 10 mA CC (Fig. 2(a)), the current varied exponentially with voltage in HRS during step 1, consistent with the inherent semiconductor conductivity of crystallized hematite. When the voltage swept across zero and reached the region between −1.0 and −1.5 V in step 2, the current abruptly jumped to the CC limit, indicating the "Set" process that facilitates conversion from HRS to LRS. In step 3, the current varied nearly linearly with voltage in LRS. When the voltage reached approximately +0.85 V in step 4, the current exhibited soft breakdown followed by a slow rise to CC (see Fig. 2(c)), indicating an incomplete "Reset" process and confirming NDR existence. Finally, conductivity reverted to HRS, completing one sweeping cycle. However, at 14 mA CC (Fig. 2(b)), the I–V curve maintained an exponential or semiconductor-like form in both HRS and LRS. The "Set" process still occurred between −1.0 and −1.5 V, but in step 4 the current dropped considerably to nearly HRS at approximately +1.2 V and nearly completely ruptured in the NDR region (see Fig. 2(d)). To further investigate current level effects, we varied the CC from 2.5 mA to 16 mA (Fig. S1, see ESI†). The results confirmed that high CC (13 mA) produces more intensive breakdown of LRS and a higher peak-to-valley current ratio. We also examined voltage range effects (Fig. S2, see ESI). At constant CC (e.g., 10 and 12 mA), nearly all curves overlapped, and the absolute voltages for the "Set" process and NDR peak were all below 2 V, indicating that voltage range has no visible effect. This work represents the first observation of RS and NDR coexistence in pure -Fe O nanorod film under external bias.

Fig. 2 I–V tests sweeping from +2 to −2 V and back to +2 V under different CC levels: (a) 10 mA and (b) 14 mA. Magnified partial I–V curves in the NDR region: (c) corresponding to 10 mA and (d) corresponding to 14 mA.

The plots of ln(I) versus ln(V) and ln(I) versus √V in [Figure 3: see original paper] were fitted to illustrate the conduction mechanisms of the I–V curves in Fig. 2. At 10 mA CC (Fig. 3(a)), the slope was 1.23 at lower absolute voltages and increased to 2.06 at higher absolute voltages in HRS for both bias polarities. This indicates that, excluding the "Set" process region, the I–V relationship in HRS initially follows metallic conduction and then transitions to space charge limited conduction (SCLC), which may be related to defects such as oxygen vacancies, dislocations, and voids.[22] The slope of the I–V curve in LRS before reaching the NDR peak voltage was 0.97, showing a nearly metallic linear trend that suggests the presence of thermally generated carriers.[22] Figs. 3(b) and 3(c) show the I–V relationships in HRS and LRS under 14 mA CC, respectively. In HRS, the logarithm of current was not linear with the logarithm of voltage but was linear with the square root of voltage, with slope values of 4.16 and 6.08 in the positive and negative bias regions, respectively. This suggests that Schottky emission, which tends to occur at interfaces aggregated with oxygen vacancies, is the dominant transport mechanism.[23,2] Under Schottky emission, carriers must cross the potential barrier via thermionic effects,[2] which is plausible under the more intensive thermal effects at high CC levels. Fig. 3(c) shows the corresponding I–V relationships in LRS. In the negative bias region, the slope

changed from 1.16 to 1.77 with increasing absolute voltage. In the positive bias region (excluding the NDR region), the slope changed from 1.09 to 1.50 and finally to 2.26 with increasing voltage. Thus, carriers always follow metallic conduction and Child' s law (SCLC) in LRS, which differs from the behavior at low CC. This further demonstrates that I-V characteristics are distinctly dependent on CC values. Based on these observations, carrier transport should be largely governed by thermal processes determined by the current level.

Fig. 3 (a) Double-logarithmic plots of I-V curves in HRS and LRS at 10 mA CC. (b) Plot of ln(I) vs. √V in HRS at 14 mA CC. (c) Plot of ln(I) vs. ln(V) in LRS at 14 mA CC.

[Figure 4: see original paper] shows the temperature dependence of resistance in HRS and LRS to further investigate carrier conduction mechanisms. We selected a CC of 14 mA, where thermal effects are sufficiently severe. In HRS (black curve), the read voltage was set at +1.4 V, and resistance decreased with increasing temperature from 150 K to 300 K, demonstrating semiconductor behavior originating from the intrinsic conductivity of hematite. This is consistent with the result shown in Fig. 2(b). In LRS, we read the resistance-temperature (RT) curves at +0.5 V and +1.4 V outside (red curve) and within the NDR region (blue curve), respectively, to distinguish differences between states before and after NDR. The red curve showed weak semiconductor behavior with a relative resistance ratio R(300 K)/R(150 K) of 0.89, whereas the blue curve exhibited an apparent transition at approximately 260 K. The semiconductor characteristic of the blue curve changed from faint to strong with an abrupt resistance increase up to 260 K, indicating a temperature-dependent change in conduction mechanism. At least two possible mechanisms may operate above 260 K, with one possibly collapsing at low temperature. The major inevitable point defects in crystallized -Fe O are oxygen vacancies and interstitial Fe^{3} or Fe^{2} ions.[2] While oxygen vacancy-induced bipolar RS in metal oxides with asymmetrical electrodes is commonly observed,[2] tunneling current may also arise from hopping conduction of electrons between multivalent ions (e.g., Fe^{3} or Fe^{2}) in metal oxides. This conduction is thermally activated and becomes localized at hopping sites as temperature decreases.[2] We propose that the synergy between oxygen vacancy migration and electron hopping between sites yields better conductivity approaching that of a conductor at temperatures above 260 K, while localization of hopping electrons below 260 K results in higher resistance.

Fig. 4 Temperature-dependent resistance (RT) curves of the film structure at 14 mA CC.

Scheme 1 illustrates the proposed mechanism involving oxygen vacancies and hopping electrons between interstitial Fe^{3} /Fe^{2} ions under external bias. Oxygen vacancies typically accumulate at heterojunction interfaces (e.g., between the oxide film and FTO substrate) due to stoichiometry mismatch and local strain.[23] At the beginning of voltage sweeping, the bottom FTO electrode was under negative bias, and positively charged oxygen vacancies tended to localize there. Only thermally generated intrinsic carriers of hematite were transported

through the film, leading to the exponential I–V relationship in HRS. After the voltage swept into the negative region, oxygen vacancies gradually migrated toward the top electrode and formed conducting filaments at the "Set" voltage. When bias returned to the positive region, these filaments ruptured slowly due to oxygen vacancies returning to the bottom electrode, causing the current to drop piecemeal and creating the NDR peak. The RT test revealed that thermal effects at room temperature were inevitable. The tunneling current formed by hopping electrons would be activated by increasing Joule heat in the conducting filaments, possibly preventing the current after the NDR peak from dropping directly to the low HRS value. However, the activation energy and hopping routes remain unclear and require future investigation. Under higher CC levels, the current dropped to nearly HRS. The compliance current could also determine the RS type because the filament current level governs the amount of Joule heat.[1],[2] Here, the gentle filament breakdown under 10 mA CC resulted from oxygen vacancy migration to the FTO bottom, while further filament fusion at 14 mA CC was caused by excessively intensive thermal effects.

Scheme 1 Schematic of the formation and rupture of conducting filaments consisting of oxygen vacancies and hopping electrons between interstitial Fe^{3} /Fe^{2} ions in the W/ -Fe O nanorod film/FTO sandwich structure.

[Figure 5: see original paper] (a) shows energetic alignments of the conduction band (E_C), valence band (E_V), and Fermi levels (E_F) of the -Fe O nanorod film, tungsten, and FTO substrates. Possible band diagrams of the W/ -Fe O and -Fe O /FTO interfaces during different processes are shown in (b) "Set" process and (c) NDR process. In Fig. 5, we discuss a possible model for the "Set" process and NDR behavior based on energy band diagrams. Fig. 5(a) summarizes the possible energetic alignments of E_C, E_V, and E_F for the -Fe O nanorod film.[33] Both interstitial metal ions and oxygen vacancies are considered donor defects in n-type semiconductor metal oxides.[3] ,[3] The formation equations for oxygen vacancies and interstitial iron ions are shown in equations (S1) and (S2) in the ESI. The donor levels (E_D) for interstitial Fe^{3} /Fe^{2} ions and oxygen vacancies are both estimated to lie between E_C and E_F. To avoid excessive carrier accumulation at the E_D level in Figs. 5(b) and 5(c), oxygen vacancies are not shown at their precise energetic locations but are arranged according to their actual positions (initially located at the interface between the -Fe O nanorod film and FTO substrate). The E_F levels of tungsten and FTO electrodes are also indicated.[3] Due to differences between the E_F levels of these three materials forming a sandwich structure, the energetic bands of hematite at both interfaces would bend until the E_F levels reached equilibrium. When a negative voltage was applied to the top W electrode (Fig. 5(b)), intrinsic electrons (major carriers) and holes (minor carriers) were driven toward the FTO substrate and W electrode, respectively, giving rise to the exponential I–V relationship of the semiconductor. Meanwhile, hopping electrons between interstitial Fe^{3} /Fe^{2} ions were driven toward the FTO substrate while positively charged oxygen vacancies were impelled toward the W electrode. These carriers caused rapid current enlargement and the final "Set" process. In

Fig. 5(c), the W electrode was under positive voltage, and all carriers reversed their direction. Under lower CC, oxygen vacancies returned to the interface between the -Fe O nanorod film and FTO substrate, causing a slight current drop in the NDR region. However, under higher CC, the intense thermal effect further destroyed the conduction contribution from hopping electrons between interstitial Fe^3 /Fe^2 ions, resulting in a drastic current drop in the NDR region.

Although I-V sweeping tests were reproducible, both the "Set" voltages and NDR peak voltages exhibited slight fluctuations (Fig. 2). This may be caused by incomplete rupture of conductive paths during NDR, which requires a variable voltage to recover the filaments during the subsequent "Set" process.[2] Fig. S3 in the ESI shows the fluctuation ranges of "Set" and NDR voltages under various CC levels. The "Set" voltage ranged from −1.81 V to −0.55 V, while the NDR voltage ranged from +0.35 V to +1.87 V. However, the retention characteristics of HRS and LRS were highly stable. [Figure 6: see original paper] shows current versus time plots for 10 min at a readout voltage of +0.5 V. Although the LRS/HRS current ratios (I_ON/I_OFF) were only 8 and 3 under 10 and 14 mA, respectively, the I–t curves remained constant throughout testing (additional I-t curves under different CC levels are provided in Fig. S4 in the ESI).

Fig. 6 Current–time curves of HRS and LRS read at +0.5 V under different CC levels: (a) 10 mA and (b) 14 mA.

Conclusions

We report for the first time the coexistence of RS and NDR in -Fe O nanorod film on FTO substrate prepared by hydrothermal methods. RS and NDR occurred under opposite voltage polarities. The I–V curves demonstrated that LRS and HRS followed different conduction models depending on current levels, which directly affected the thermal process. We attribute the observed phenomenon to defects (oxygen vacancies, interstitial iron ions, etc.) in the -Fe O nanorod film. This work provides valuable information on the electrical characteristics of -Fe O nanorod films for the low-cost production of efficient switching devices.

Acknowledgements

This work was financially supported by the National Basic Research Program of China (2014CB931704), the National Natural Science Foundation of China (NSFC, Nos. 51371166, 51571186, 11404338, 11304315, 51401206, 11504375), and the China Postdoctoral Science Foundation.

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